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Actuation response of polyacrylate dielectric elastomers

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ABSTRACT

Polyacrylate dielectric elastomers have yielded extremely large strain and elastic energy density suggesting that they are useful for many actuator applications. A thorough understanding of the physics underlying the mechanism of the observed response to an electric field can help develop improved actuators. The response is believed to be due to Maxwell stress, a second order dependence of the stress upon applied electric field. Based on this supposition, an equation relating the applied voltage to the measured force from an actuator was derived. Experimental data fit with the expected behavior, though there are discrepancies. Further analysis suggests that these arise mostly from imperfect manufacture of the actuators, though there is a small contribution from an explicitly electrostrictive behavior of the acrylic adhesive. Measurements of the dielectric constant of stretched polymer reveal that the dielectric constant drops, when the polymer is strained, indicating the existence of a small electrostrictive effect. Finally, measurements of the electric breakdown field were made. These also show a dependence upon the strain. In the unstrained state the breakdown field is 20 MV/m, which grows to 218MV/m at 500% x 500% strain. This large increase could prove to be of importance in actuator design.

Keywords: dielectric, elastomer, adhesive, strain, actuator, electrostriction, electric breakdown

1. INTRODUCTION

Actuators made from materials that deform in response to applied electric fields have been a promising area of research for several years now [1-11]. Progress is still being made. Giant strain has recently been obtained from dielectric elastomer actuators [11-12]. Even though very good results have been obtained, many aspects of the underlying physics needs to be understood. Much work is needed on the basic understanding of the performance of a given dielectric elastomer actuator with respect to materials research, as well as the effect of the specific geometric configuration. Hopefully, this understanding will guide researchers in choosing and developing better materials for later generations of the dielectric elastomer actuators. In this paper we focus on understanding the basic physical mechanism underlying the observed electromechanical response. Because the maximum performance will depend on the maximum electric field that can be applied to the material, we also consider factors affecting the dielectric strength of the material.

As a case study we focus on elastomers derived from the very high bond structural adhesive from 3M™, sold as VHB™ 4910. This material is deserving of interest since it has produced the greatest strains and elastic energy density of any known dielectric elastomer material [12]. VHB™ 4910 is a pressure sensitive adhesive elastomeric film. It is a closed cell, chemically cross-linked amorphous polyacrylate network [13]. This material is available as a pre-cast 1mm thick polyacrylate foam adhesive. The film is translucent, and extremely compliant. It can stretch at least 36 times in area size without breaking. Finally it is very easy to work with, as no additional curing is required.

2. THEORETICAL

A dielectric elastomer actuator can be considered as a compliant capacitor. The electrodes (capacitor plates) are made such that they can greatly change their size, without losing their conductivity. The medium consists of a thin layer of insulating, dielectric elastomer, the basic element is shown in Figure 1.

A pressure arises between the two electrodes of a capacitor when it is charged. This pressure, known as the Maxwell pressure, arises from the fact that the plus-charges on one electrode attract the minus-charges on the other. If the electrodes and the medium are compliant, the result for the generated Maxwell pressure is

$$p = \varepsilon\varepsilon_0 E^2 , \qquad (1)$$

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where ε is the relative dielectric constant of the medium, ε_0 is the vacuum permittivity, and E is the applied electric field [8-11]. This equation is valid assuming that the relative dielectric constant is not a function of the applied electric field or the stress or strain in the material. These assumptions may be experimentally verified. First it is necessary to relate the parameters in equation 1 to physically measurable quantities.

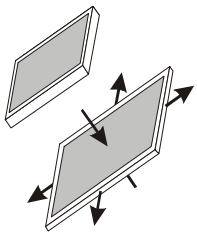

Figure 1: Principle of Maxwell pressure. When a voltage between the compliant electrodes is applied a pressure arises, and the elastomer compresses while it expands in the plane.

Consider a film of elastomer with an initial thickness z_0 and a final (equilibrium) thickness z. Accordingly, the length of the film is y_0 and y, and the width is x_0 and x. The pressure, p, is applied along the z-direction. To test the validity of equation 1, we will need to calculate the resulting pressures and forces along the x and y-axis. These physically measurable quantities may be found from the following considerations.

The infinitesimal work, dW, generated when a film plane is moved against a force, $\vec{F}$, an infinitesimal distance, $d\vec{x}$, is

$$dW = \vec{F} \cdot d\vec{x} = F_x dx + F_y dy + F_z dz .$$

At a stable equilibrium (when there is no motion of the film) $dW = 0$, so the above equation may be rearranged to

$$F_z dz = -F_x dx - F_y dy$$

Both sides of the equation are divided by dz, resulting in two derivatives, which are evaluated by noting that the volume is constant, $V = xyz$,

$$F_z = -F_x \frac{dx}{dz} - F_y \frac{dy}{dz} = F_x \frac{x}{z} + F_y \frac{y}{z} .$$

Both sides are now divided by the area $A_z = xy$ of the side of the film which is perpendicular to the z-direction

$$\frac{F_z}{xy} = \frac{F_x}{yz} + \frac{F_y}{zx},$$

which by definition implies that $\sigma_z = \sigma_x + \sigma_y$. A special case of the above equation is when the width of the film is kept constant, $dx = 0$, then the above derivation reduces to

$$\sigma_z = \sigma_y . \qquad (2)$$

This relation makes it possible to relate the Maxwell pressure generated in the z-direction with the measured force in the y-direction.

The polymer film is stretched α_x times in the width direction, and α_y times in the length direction, resulting in a film with a final thickness $z = z_0 / (\alpha_x \alpha_y)$. The final length of the actuator is $y = \alpha_y y_0$. The relation between the applied voltage, V, and the force measured on the force gauge, F_y, can be obtained from (2), $\sigma_z = \sigma_y = F_y / A_y$. Inserting all the variables results in the relation

$$F_y = \frac{x_0 z_0}{\alpha_y} \varepsilon \varepsilon_0 E^2 = \frac{x_0}{z_0} \alpha_x{}^2 \alpha_y \varepsilon \varepsilon_0 V^2 . \qquad (3)$$

3. EXPERIMENTAL

Equation 3, for evaluating the field-induced stress, was evaluated experimentally for the elastomer VHB™ 4910 from 3M™. The material is a clear and colorless film, with a dielectric constant of 4.7 in the unstretched state. It is very sticky and can be stretched very far in both directions in the plane of the film. It is supplied as a 1-mm-thick sheet on a half meter wide roll, mounted on a red plastic liner, which makes it very easy to cut to the preferred size. Due to its stickiness it is easy to stretch and mount on frames, making it a very good material for testing and demonstration devices.

3.1 Maxwell stress

The geometry of the setup for measuring the field-induced stress is shown in figure 2. One of the beams is fixed to a translation table, while the other is attached to a force gauge using a piece of string, making it impossible for the elastomeric film to change in length. This is known as a ‘constant strain’ experiment, and the force measured on the force gauge, arising from the applied voltage, is known as the blocking force. This experiment duplicates the equilibrium condition used to derive equation (3).

A piece of VHB 4910 was pre-stretched on a frame, 6.0 times (500% pre-strain) in the *x* direction and 4.0 times (300% pre-strain) in the *y* direction. Plastic beams 200 mm long and 10 mm wide were glued to the film (on both sides for symmetry) along the 500% pre-strain direction, 20 mm apart along the 300% pre-strain direction. Thin strips of VHB 4910 were attached to the film on the edge of the area spanned by the beams (on both sides for symmetry), in order to contain any cracks originating on the edge of the film. The strips also keep the active area relatively square when the actuator is cut from the frame. CircuitWorks™ conductive carbon grease was smeared on both sides of the film, and aluminium foil connectors were stuck to each side. String was attached to the beams, and the actuator was mounted in series with a force gauge. Alligator clamps were attached to the wires, such that they did not add varying weight to the setup. The pre-strain in the length direction was controlled by a micrometer-screw. When the desired length was reached, the force-gauge was zeroed, thus negating force due to the weight and spring force of the actuator. Voltage was applied using a high-voltage DC-source. The voltage at the equilibrium condition was measured together with the corresponding force measured on the force-gauge.

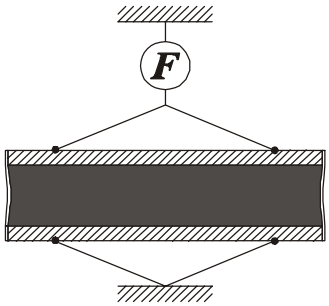

Figure 2: 200 mm wide VHB™ 4910 film, in a constant strain measurement setup. The *x* and *y* directions are horizontal and vertical, respectively.

The maximum applied voltage is determined experimentally as the voltage at which the film starts to wrinkle, the ripples are in the width direction of the sample. At higher voltages the force is constant on the force gauge, because all the extra force is guided into the width direction. Equilibrium in this direction is apparently reached by allowing the film to expand and wrinkle in this direction. If the voltage is increased further above this limit, mechanical breakdown in the film will occur, therefore the wrinkle limit was chosen as the maximum for applied voltage. For all three pre-strains the voltage wrinkle limit was the same, typically about 5.0 kV, but the force generated at this voltage was different, see figure 4. At the lowest pre-strain (300% in the length direction), the max force is ~ 470 g, while at 500% it is ~ 670 g.

Measurements were made for pre-strains of 300%, 400%, and 500%. The results for 300% are shown in figure 3. The measured voltage and developed force are plotted on the secondary axes, and calculated electric field vs. stress is plotted on the direct axes. The dotted lines are the expected parabolic curves corresponding to the relative dielectric constant written next to the curve. The experimental curve follows a parabolic behavior very nicely, with an *apparent* relative dielectric constant of approximately 4.0.

In the insert of figure 4, the stress vs. electric field curve is plotted for the three pre-strains. They all fall on the same 'master' curve, the parabolic curve predicted by (3), though there is a slight tendency for the higher pre-strain curves to produce slightly less force in response to the applied voltage. The same measurements were repeated with other samples,

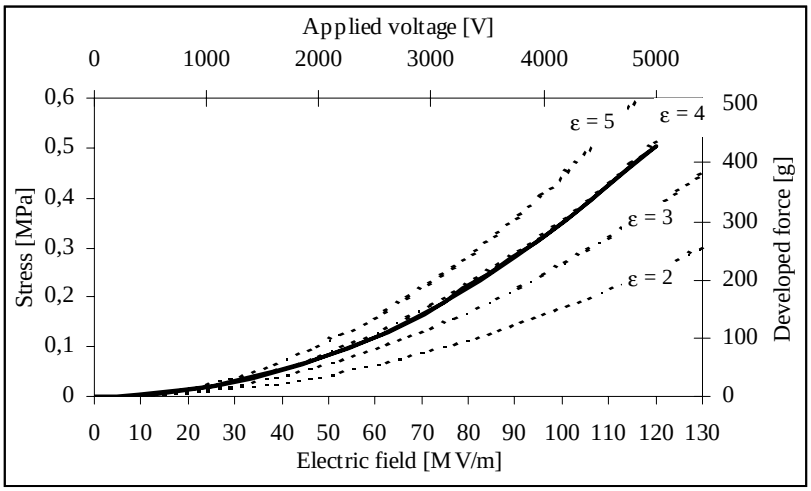

Figure 3: Stress/force vs electric field/voltage curve for sample with 500% x 300% pre-strain.

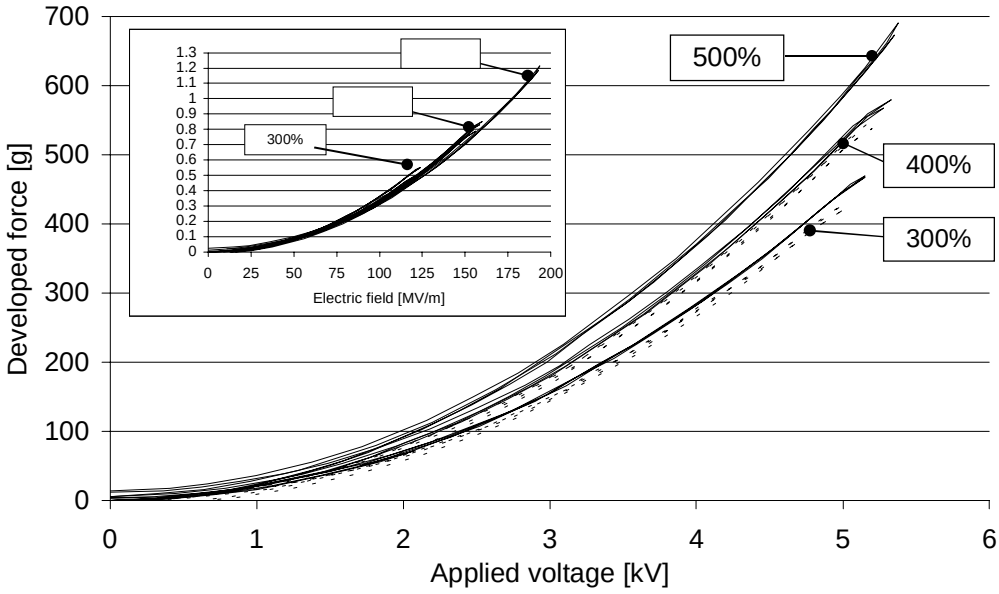

Figure 4: Force-voltage curve for the same sample at different strains in the length direction.

and the same behavior was found, thus it is verified that the generated force has a parabolic dependence upon the electric field, as predicted by (3).

The dielectric constant of the unstrained material is 4.7 according to 3M™ literature [13]. The 18% lower performance of the actuator might be explained by deviations between the test set up of figure 2 and the ideal rectangular, uniform thickness film assumed in equation (3). The deviations are mainly present on the edges of the film. The 500% prestrain in the width direction will tend to pull the edges inwards, thus lowering the area, and adding locally to the thickness of the film, which all results in lowered performance of the actuator. Still, the lower performance might also be explained by a lower dielectric constant, which could come about as a consequence of the high strain. Direct measurements on the relative dielectric constant are discussed in the next section.

3.2 Dielectric constant of stretched VHB™ 4910

The measurements of the blocking force of the dielectric actuator provided evidence to support the description based upon equation (3). Unfortunately, the measured stress was systematically lower than was predicted by (3), using the dielectric constant stated by the manufacturer for VHB™ 4910. Inspection of the capacitance of the actuator with a simple multimeter also showed that it was lower than what could be inferred from a simple evaluation using the common formula $C = \varepsilon\varepsilon_0 A / d$. Therefore it was deemed necessary to explicitly investigate the dielectric constant of the stretched material.

The film was pre-strained on a frame, a series of pre-strains were made, from 0%x0% to 400%x400%, in increments of 100%, where the percentages indicate the strain in each of the two planar film dimensions. Plastic rings with an inner diameter of 30 mm were placed juxtaposed on both sides of the film. Each side of the film was brushed thoroughly with carbon black for good contact. Planar metal electrodes with a diameter of 30 mm were placed on each side of the film, these act as electrodes in conjunction with the carbon black.

The sample was then inserted into a Novocontrol™ Dielectric Analyzer, an extremely sensitive dielectric spectrometer. The dielectric spectrometer measures the impedance of any sample at a given frequency, and with software analysis it was possible to extract information on material parameters such as the dielectric constant and loss factor. The dielectric spectrometers frequency range lies from $3 \cdot 10^{-5}$ Hz to $2 \cdot 10^{7}$ Hz, here we only scanned from $1 \cdot 10^{-1}$ to $1 \cdot 10^{6}$ Hz. The dielectric constant was extracted from the real part of the impedance for frequencies 0.1, 1, 10 and 100 Hz, and plotted versus the area increase ratio (see figure 5). The area increase ratio is defined as

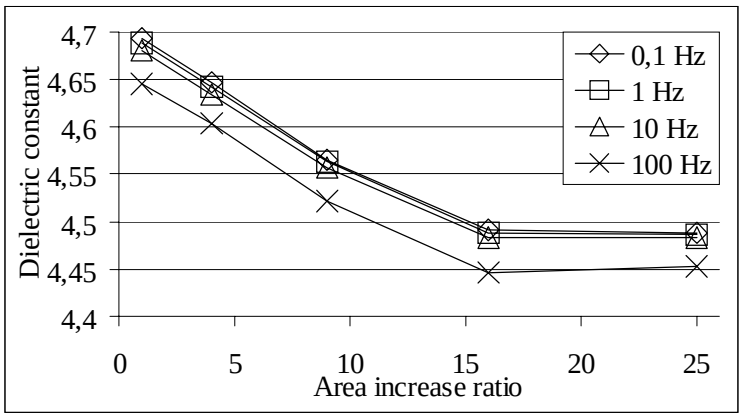

Figure 5: The relative dielectric constant of VHB™ 4910 drops, when it is stretched.

$A/A_0 = \frac{1}{(1+\alpha_x)(1+\alpha_y)}$,

and is inversely proportional to the thickness of the film after it has been stretched.

These measurements reveal a tendency of the real part of the dielectric constant to drop when the film is stretched. A possible reason for the drop is the fact that a strained polymer allows less movement of its constituent polar segments. As dielectricity arises from the affinity that polar molecules have for aligning themselves with an external electric field, it is plausible that straining the polymer lowers the polar segment reorientation, and thus lowers the dielectricity (or dielectric constant).

This variation in the dielectric constant with strain suggests that other mechanisms beyond Maxwell stress may be contributing to the observed response. A stress or strain induced in a dielectric in response to a change in a materials dielectric constant due to a variation in strain is known as electrostriction [6,15]. This stress is also second order in the electric field, thus the simplest formula incorporating this effect can be written on the form

$$p = \varepsilon\varepsilon_0(1-\gamma)E^2, \quad (4)$$

which is essentially the same as equation (1), only with a scaling parameter γ, denoting the effect of electrostriction. An expression for γ, also called the electrostriction coefficient [1], may be derived using an approach similar to that described in [9]. This analysis yields

$$\gamma = 0.5\frac{zd\varepsilon}{\varepsilon dz} = -0.5\frac{Ad\varepsilon}{\varepsilon dA}. \quad (5)$$

Estimates of this coefficient can be made from the data in Figure 5. Although there are too few data points to make an accurate assessment of the value near the 300% pre-strain point, a worst case (largest value) is 0.018. The electrostriction in this case is an *unwanted* property, as it *subtracts* from the efficiency of the actuator. This is in contrast to the situation in other polymer films [7], where the electrostriction *adds* to the ordinary Maxwell pressure of any capacitor.

The value for γ indicates that electrostriction reduces the expected response of equation 1 by less than 2%. As it appears, the weakly electrostrictive behavior of VHB™ 4910 is not enough to explain the lower performance of the material. We then suggest that the lowered performance is primarily due to a discrepancy between the 'theoretical' geometry of equation (3), and its actual realization as discussed above. These discrepancies are probably due to the fact that the edges of the actuator are not straight, but curved inwards toward the center of the actuator. The consequences of this are both that the active area is reduced, and that the thickness of the actuator increases locally, thus effectively reducing the electric field. Both these effects would tend to reduce the measured force of equation (3).

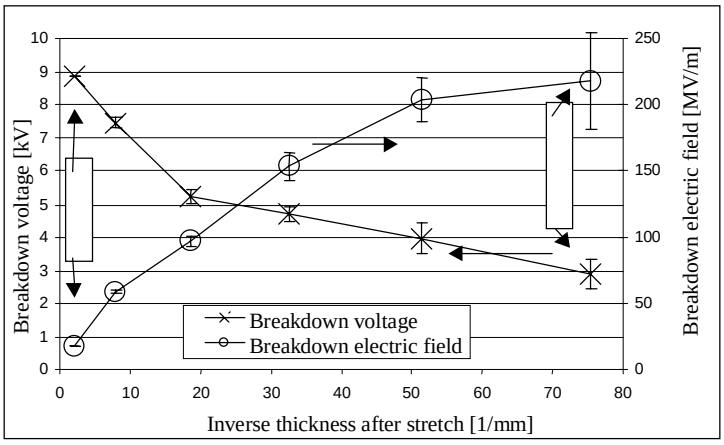

Figure 6: Electrical breakdown strength of isotropically pre-strained VHB™ 4910. The film with a pre-strain of eg. 300 % x 300 % has increased (1+ 300%)x(1+ 300%)=16 times in area.

3.3 Electric breakdown field

The films made from VHB™ 4910 are often driven up to voltages of 5 kV and higher. The resulting electrical field ranges from 50 MV/m and up, depending upon the thickness of the film after it has been pre-strained. With the advent of the switch-mode amplifier, it is possible to build very small electrical circuits that can supply these high voltages. Therefore the limit for high voltage actuators is not the high voltage that is available from the power supply, but rather the electric breakdown field, which is the electric field at which a given dielectric material experiences a finite conductivity due to charges generated in the material. For VHB™ 4910 material the electric breakdown is fatal, thus when an actuator 'burns', it is beyond repair. Therefore it is important to know the limitations on the applied voltage.

A pair of 0.25 inch diameter brass electrodes where clamped around a film such that the distance between them could be read on a Mitotuyo™ thickness gage. The voltage supply was connected to these electrodes. Samples were pre-strained the same amount in both directions in the plane, from 0% to 500% in increments of 100%. The film was inserted between the clamps, and the thickness was noted. The voltage was increased slowly until electrical breakdown occurred. About 30 measurements were made for the same pre-strain. The results for these *isotropic* pre-strains (same pre-strain in both directions) are plotted in figure 6.

It is important to point out that the volume was conserved, in that the final thickness predicted from the strain ratio, matched the measured thickness of the samples. Also, during application of high voltage, the thickness of the sample between clamps did not change, which is indication that the volume of the sample is conserved even during actuation.

The interesting material dependent property here is the *electric field* at breakdown. One theory for the breakdown electric field says that when the electric field is high enough, an otherwise stable space charge is accelerated, thus creating a catastrophic avalanche spark [14], very much like in a Geiger-Müller tube. In VHB™ 4910 this breakdown event is fatal for two reasons: the volume around the spark becomes conducting, and cracks from the spark volume very easily propagate causing the whole film to rip. Thus it is important to find methods to improve the breakdown strength of the elastomeric medium. One such possible method is to pre-strain the elastomer, as is signified by the experimental data of figure 6. In going from a pre-strain configuration of 0% in both directions in the plane to 500%, the electrical breakdown strength increases from 18 MV/m to 218 MV/m. These results are consistent with those reported in Pelrine et al. [11]. We argue that the breakdown strength across a polymer chain is higher than along it, due to a higher collision cross section between avalanching electrons and polymer atoms. Therefore the increase in breakdown strength can be ascribed to the orientation of the polymer chains in the plane of the film due to the pre-strain, and perpendicularly to the electric field lines.

4. CONCLUSION

An equation connecting the theoretical expression for the Maxwell pressure with the measurable properties in an experimental setup was derived. The experiment was then performed using VHB™ 4910 acrylic adhesive as the dielectric elastomer, and carbon grease as electrodes. These experiments support the theoretical analysis, in showing a parabolic dependence of the force upon applied voltage, but the generated force was systematically too low. The discrepancy was ascribed to imperfection in experimental setup geometry and a small contribution from a possible electrostrictive effect. The size of the electrostrictive behavior is determined from dielectric spectroscopy performed on strained films, and shown to be smaller than is needed to completely explain the diminished performance of the setup, so we conclude that almost all of the discrepancy is due to imperfections in the setup.

The electric breakdown field was also investigated, and it was shown to be highly dependent upon the strain. It was then concluded that this would ultimately be of consequence in actuator design. Further research should look at other types of dielectric elastomer materials and other effects such as viscoelasticity and electrical loss factors in an attempt to further understand the electromechanical behavior of dielectric elastomer materials.

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